

COMPARATIVE STUDY OF FLUX PINNING, CREEP AND CRITICAL CURRENTS BETWEEN  
YBaCuO CRYSTALS WITH AND WITHOUT  $Y_2BaCuO_5$  INCLUSIONS

Masato MURAKAMI, Satoshi GOTOH, Hiroyuki FUJIMOTO, Naoki KOSHIZUKA  
and Shoji TANAKA  
International Superconductivity Technology Center,  
Superconductivity Research Laboratory,  
1-10-13 Shinonome, Koto-ku, Tokyo 135 JAPAN

**ABSTRACT** It is possible to distribute fine  $Y_2BaCuO_5$  inclusions into  $YBa_2Cu_3O_{7-x}$  crystals by a melt process utilizing a peritectic reaction:  $Y_2BaCuO_5 + L \rightleftharpoons YBa_2Cu_3O_{7-x}$ . We prepared Y-Ba-Cu-O crystals with and without  $Y_2BaCuO_5(211)_2$  inclusions and compared critical currents and flux creep. It was found that the crystal with fine 211 inclusions exhibit much higher  $J_c$  values and lower flux creep rate than the crystal without 211 inclusions. This result indicates that fine dispersion of 211 inclusions can contribute to flux pinning as in the case of conventional superconductors.

## 1. INTRODUCTION

In type II superconductors, the introduction of effective pinning centers is required in order to obtain high critical current density ( $J_c$ ). However, in the case of high  $T_c$  oxide superconductors, it is not yet clear whether the conventional pinning theory can explain their critical current characteristics or not. It is also believed that extremely small coherence length and high anisotropic nature in electric conduction may play a dominant role in flux pinning. However, it has been reported that the introduction of pinning centers either through neutron irradiation<sup>1,2)</sup> or metallurgical process<sup>3)</sup> can help increase critical current density.

We have recently found a novel melt processing technique which enables us to fabricate large Y-Ba-Cu-O crystals with various volume fractions of  $Y_2BaCuO_5$  inclusions<sup>4)</sup>. It is known that non-superconducting inclusions can work as effective pinning centers in conventional superconductors<sup>5)</sup>. They are also expected to contribute to flux pinning in oxide superconductors.

In this study, we prepared two kinds of Y-Ba-Cu-O crystals with and without  $Y_2BaCuO_5$  inclusions by the Melt-Powder-Melt-Growth (MPMG) process. Then we compared critical currents and flux creep between them in order to clarify if fine dispersion of 211 inclusions is beneficial to flux pinning.

## 2. EXPERIMENTAL

### 2.1. Sample preparation - MPMG process<sup>4)</sup>

Y-Ba-Cu-O powders with nominal compositions of  $YBa_2Cu_3O_{7-x}$  and  $Y_{1.40}Ba_{2.2}Cu_{3.2}O_y$  were prepared by mixing  $Y_2O_3$ ,  $BaCO_3$  and  $CuO$  and calcining at  $950^\circ C$  for 3.2 h. Then they were subjected to the MPMG process schematically illustrated in Fig. 1. The powders were heated to temperatures above  $1300^\circ C$  and held until the powders reacted and melted completely in platinum crucibles. The melt was cooled rapidly by quenching using cold copper plates. The quenched plates

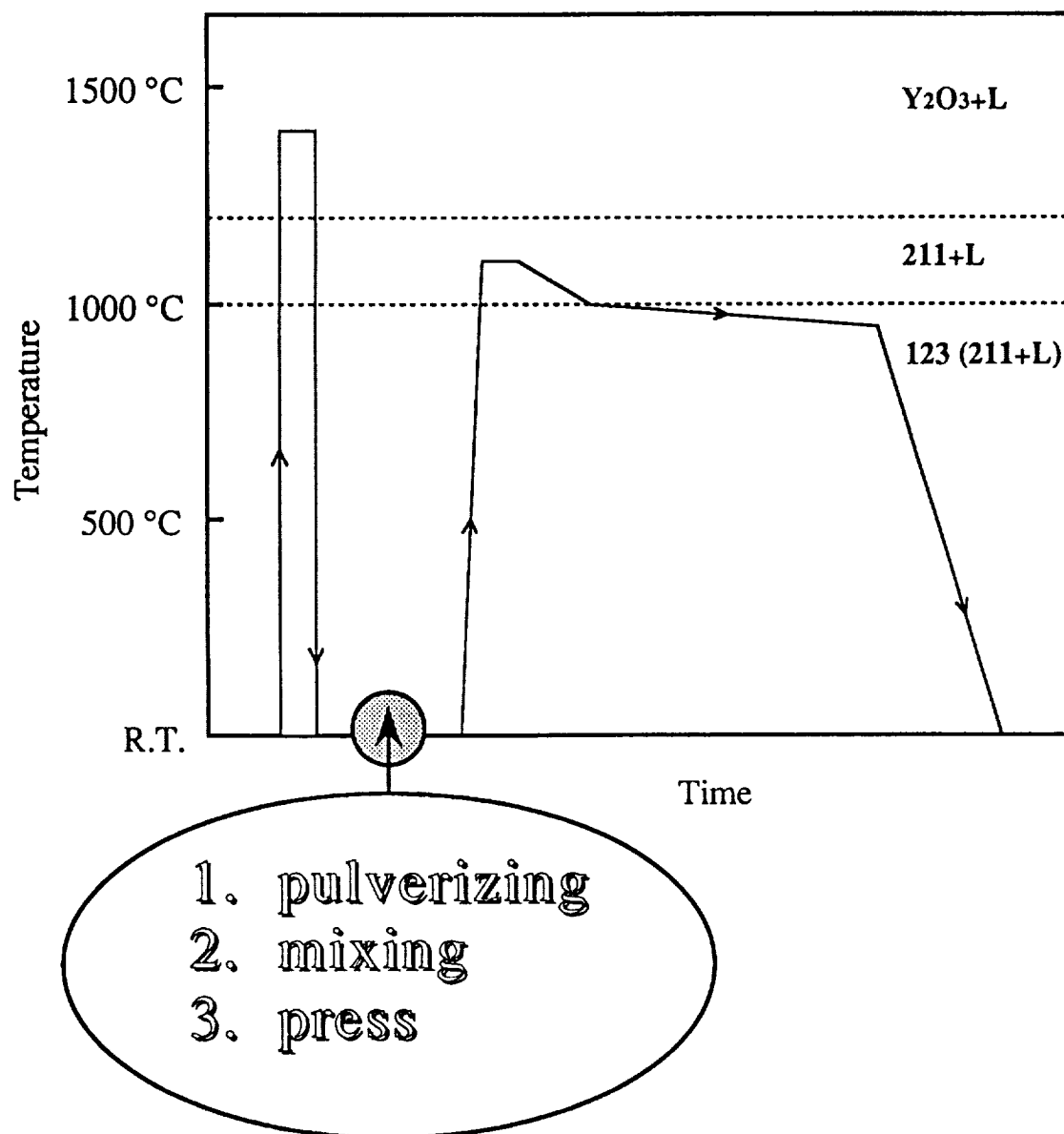


Fig. 1. Schematic illustration for Melt-Powder-Melt-Growth (MPMG) process. Y-Ba-Cu-O calcined powders are heated to temperatures above  $1300^{\circ}\text{C}$  where  $\text{Y}_2\text{O}_3$  and liquid are stable. The melt is cooled down to room temperature then ground and mixed well in order to refine and homogenize the distribution of  $\text{Y}_2\text{O}_3$ . The melt powders are pressed into various shapes such as pellets and then reheated to around  $1100^{\circ}\text{C}$  where  $\text{Y}_2\text{BaCuO}_5$  (211) and liquid are stable. At this stage 211 nucleates from  $\text{Y}_2\text{O}_3$  as fine whiskers. The sample is cooled down to  $1000^{\circ}\text{C}$  followed by slow cooling. During the slow cooling below  $1000^{\circ}\text{C}$ ,  $\text{YBa}_2\text{Cu}_3\text{O}_7$  phase nucleates and grows continuously. When we start with the compositions off the stoichiometry toward 211 rich region, fine dispersion of 211 phase is possible in the 123 matrix. After the process, the sample is annealed at  $600^{\circ}\text{C}$  for 1h and furnace cooled in flowing oxygen to incorporate oxygen into the sample.

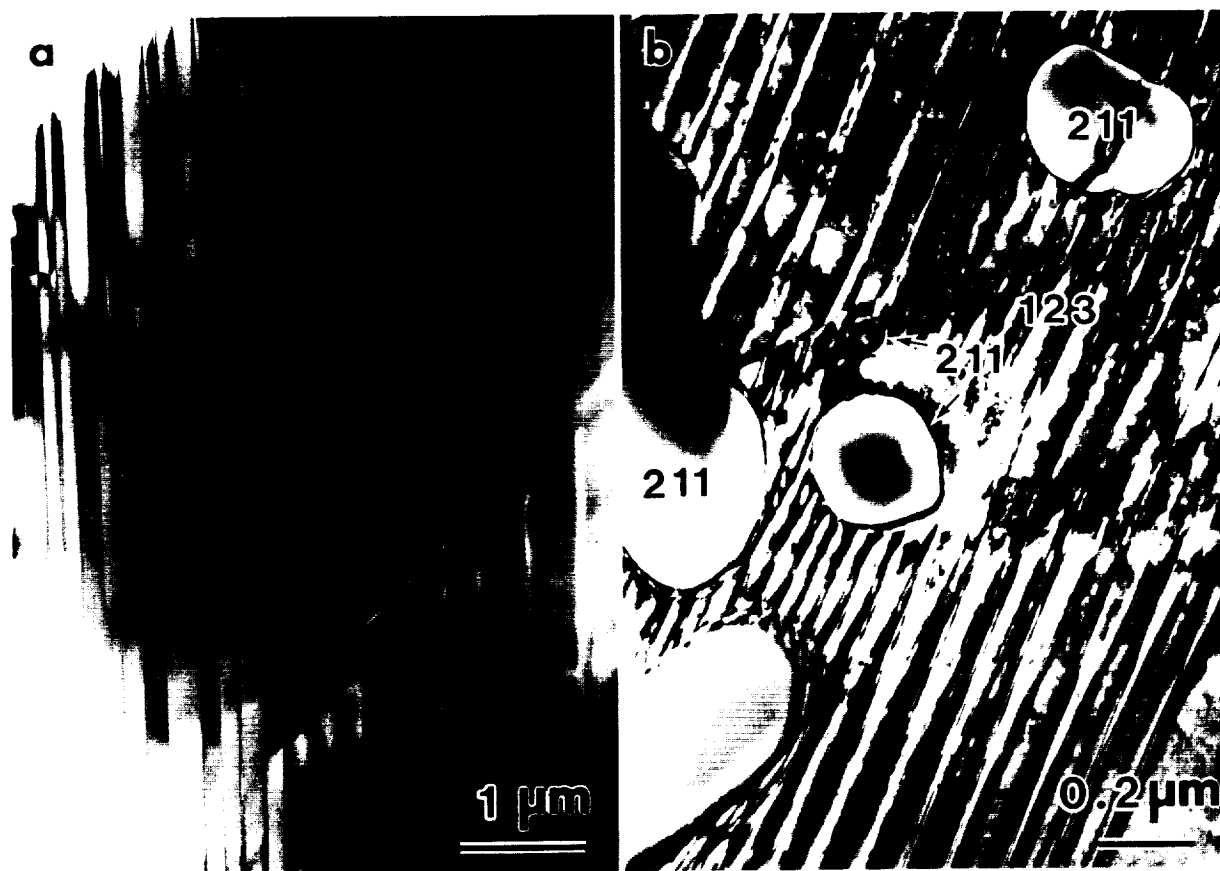


Fig. 2. Transmission electron micrographs for microstructures of (a)  $\text{YBa}_2\text{Cu}_3\text{O}_7$  crystal and (b)  $\text{Y}_{1.4}\text{Ba}_{2.2}\text{Cu}_{3.2}\text{O}_y$  crystals.

were then ground to powders about  $1\text{ }\mu\text{m}$  in diameter and mixed well and pressed into pellets  $2\text{ cm}$  in diameter and  $5\text{ mm}$  in thickness. The pellets were again heated to  $1100^\circ\text{C}$  for  $20\text{ min}$  and cooled down to  $1000^\circ\text{C}$  at a rate of  $100^\circ\text{C/h}$  followed by slow cooling at a rate of  $1^\circ\text{C/h}$  down to  $950^\circ\text{C}$  and furnace cool. The whole process was done in air. Finally the pellets were annealed at  $600^\circ\text{C}$  for  $1\text{ h}$  and slow cooled in flowing oxygen to incorporate oxygen into the samples. Fig. 2 shows transmission electron micrographs for the Y-Ba-Cu-O crystals with 1:2:3 and 1.4:2.2:3.2 compositions. It is noticeable that the former does not contain 211 inclusions, while the latter contain about 20% volume fraction of fine 211 inclusions.

## 2.2 Physical properties

Critical current densities were obtained from magnetization loops by using the following relations<sup>6,7)</sup>:

$$4\pi(M^+ - M^-) = (4\pi/10) J_c(d/2) \quad (1)$$

where  $M^+$  and  $M^-$  are magnetization in the increasing and decreasing field processes in  $\text{emu/cm}^3$ ,  $J_c$  is critical current density in  $\text{A/cm}^2$  and  $d$  is sample thickness in  $\text{cm}$ . The samples were cut from MPMG processed pellets into thin plate of  $2 \times 2 \times 0.4\text{ mm}^3$  so that the  $c$  axis was perpendicular to the surface plane. Magnetic field was applied parallel to the surface plane and therefore

perpendicular to the c axis.

Flux creep measurements were conducted with monitoring time decay of magnetization<sup>8,9)</sup>. The pinning energy U was obtained according to the following relation:

$$dM(t)/M_0 = -(kT/U) dlnt \quad (2)$$

where  $M_0$  is the initial magnetization, t is time, k is Boltzmann constant and T is temperature.

### 3. EXPERIMENTAL RESULTS

#### 3.1. Magnetization measurements

Fig. 3 shows magnetization loops for Y-Ba-Cu-O crystals with and without 211 inclusions. It is clear that the crystal with 211 inclusions exhibits much larger magnetic hysteresis and therefore larger  $J_c$  values. Fig. 4 shows magnetic field dependence of  $J_c$  for these two crystals obtained from magnetization results.

#### 3.2. Flux creep measurements

Fig. 5 shows time decay of magnetization for the Y-Ba-Cu-O crystals under magnetic fields of 1kOe and 5kOe at 77K. The relaxation takes place logarithmically with time as shown in the figure. It is also clear that the crystal with 211 inclusions with higher critical current shows lower flux creep rate or higher pinning energy. Fig. 6 shows magnetic field dependence of the pinning energy U for the crystals at 77K obtained using the relation (2). The pinning energy scales with  $B^{-1/2}$  in the both samples. These results support that the fine dispersion of 211 inclusions into the 123 matrix can contribute to flux pinning.

### 4. DISCUSSION

#### 4.1. Theoretical estimation of $J_c$ <sup>10,11)</sup>

It is known that non-superconducting phase can work as a strong pinning center in conventional superconductors. When the size of pinning center is d, the elementary pinning energy  $U_p$  is obtained as

$$U_p = (H_c^2 / 8 \pi) \pi \xi^2 d \quad (3)$$

where  $H_c$  is the thermodynamical critical magnetic field,  $\xi$  is Ginzburg-Landau coherence length. Then the elementary pinning force  $f_p$  is given by

$$f_p = U_p / 2 \xi = (H_c^2 / 16) \xi d \quad (4)$$

In order to obtain bulk pinning force  $F_p$  per unit volume, we need to sum up  $f_p$  considering the contribution of various pinning centers. In the case of non-superconducting inclusions with density  $N_p$  and d in size.  $F_p$  is obtained as

$$F_p = (N_p d / a_f) f_p \quad (5)$$

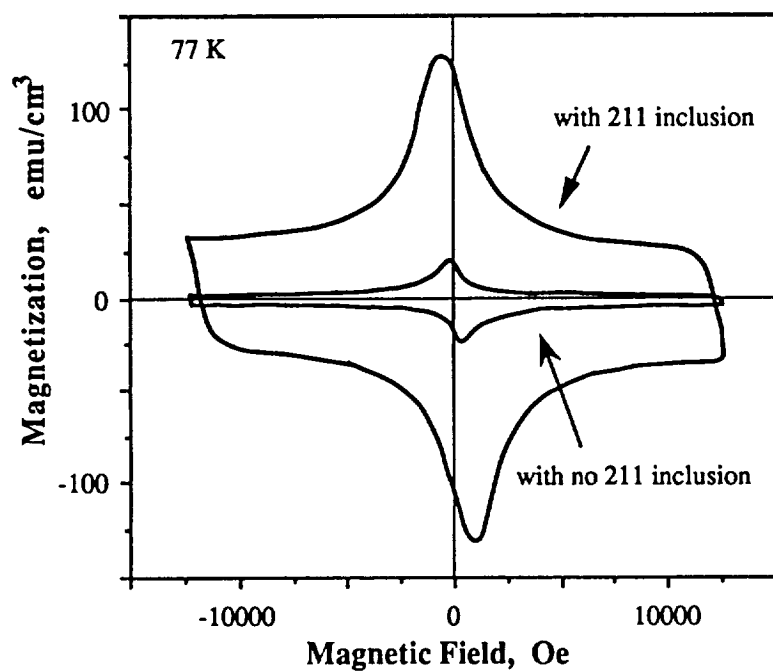


Fig. 3. Magnetization loops for the Y-Ba-Cu-O crystals with and without 211 inclusions. Note that the crystal with 211 inclusions exhibits larger hysteresis.

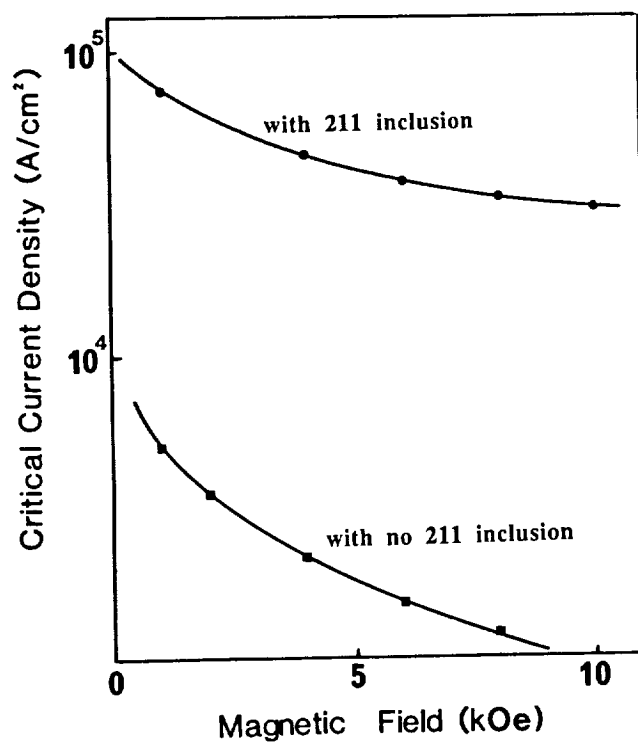


Fig. 4. Magnetic field dependence of  $J_c$  for the Y-Ba-Cu-O crystals with and without 211 inclusions. Note that the crystal with 211 inclusions exhibits much higher  $J_c$  values.

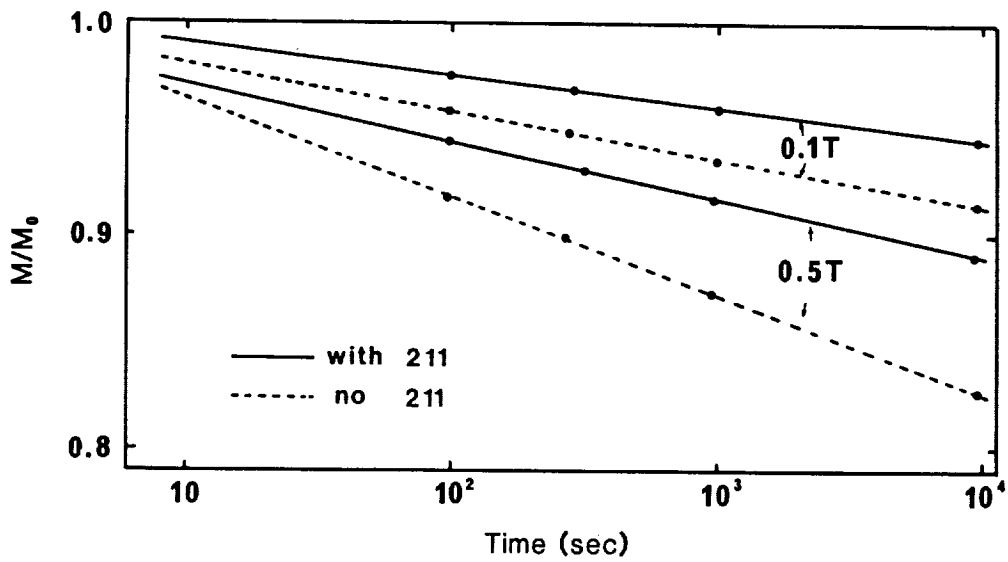


Fig. 5. Time decay of magnetization for the Y-Ba-Cu-O crystals with and without 211 inclusions under magnetic field of 1kOe and 5kOe at 77K. Note that the flux creep rate is smaller in the crystal with 211 inclusions exhibiting higher  $J_c$  values. This result indicates that the flux creep rate can be reduced by increasing  $J_c$  or the pinning force.

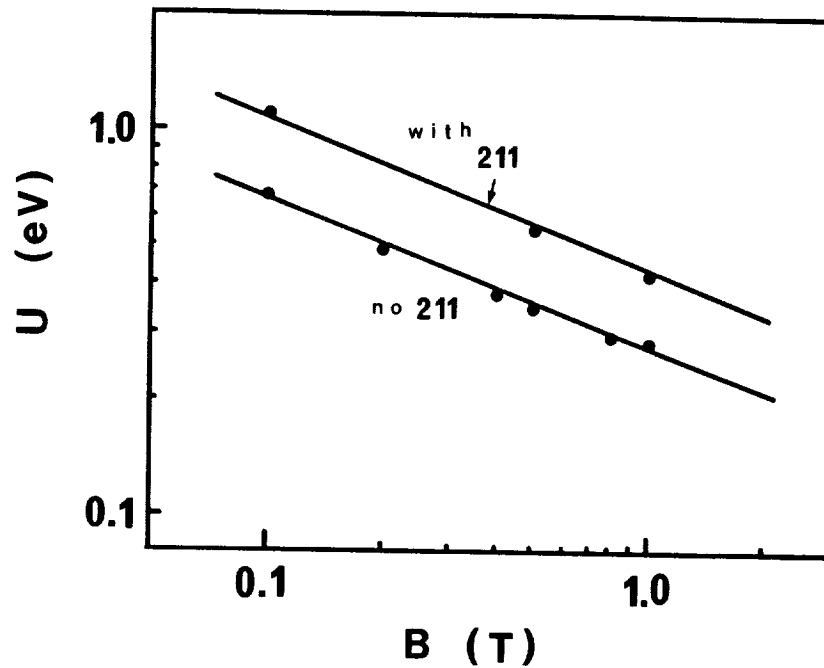


Fig. 6. Magnetic field dependence for the pinning energy  $U$  in the Y-Ba-Cu-O crystals with and without 211 inclusions.  $U$  scales with  $B^{-1/2}$  in the both samples.

where  $a_f$  is the flux lattice spacing and given by  $a_f = (\phi_0/B)^{1/2}$ . On the other hand,  $J_c$  is obtained from the relation  $F_p = J_c B/10$  and therefore a  $J_c$  value expected from the contribution of 211 inclusions is obtained as

$$J_c = 10 F_p / B = (10 N_p d_p^2 / a_f) (H_c^2 \xi / 16 B) \quad (6)$$

In the case of the crystal with 211 inclusions studied in this paper, the volume fraction of 211 inclusions is about 0.2 and the average 211 particle diameter is about  $2.5 \mu\text{m}$ . For the estimation of  $J_c$  we used  $H_c$  value of 30000e and the coherence length of 20 A at 77K. Then we obtain a  $J_c$  value of  $35000 \text{ A/cm}^2$  at 77K and 10kOe, which is in good agreement with the experimental results.

The fact that the crystal without 211 inclusion exhibit a  $J_c$  value of order  $10^3 \text{ A/cm}^2$  indicates that other defects such as twin planes and dislocations can also contribute to flux pinning, although the effect is much smaller than the contribution of 211 inclusions.

#### 4.2. Flux pinning and creep

According to the thermally activated flux creep model<sup>12)</sup>, flux creep rate  $v$  is given by

$$v = v_0 \exp(-E/kT) \quad (7)$$

where  $v_0$  is the flux creep rate when no pinning barrier exists and  $E$  is an effective pinning barrier and given as  $E = U - J_c B V x$ .  $U$  is the pinning energy,  $V$  is the activation volume of flux bundle which hops collectively and  $x$  is the distance for the bundle to move.  $J_c$  or magnetization  $M$  is considered to decay with time because of flux creep. Time relaxation of  $M(t)$  is given by relation (2). Experimental data seem to follow logarithmic time dependence. In the critical state, the effective barrier becomes zero, and therefore the pinning energy  $U$  is obtained as

$$U = J_c B V x \quad (8)$$

$V$  and  $x$  are closely related to the stiffness of flux line lattice and depend on  $B$ . According to Matsushita's analysis<sup>13)</sup>

$$U = \text{const. } J_c^{1/2} B^{-1/4} \quad (9)$$

This relation indicates that the pinning energy can be increased with increasing  $J_c$ , which is consistent with the present results.

It is also found from Fig. 4 that  $J_c$  scales with  $B^{-1/2}$  in MPMG processed crystals. From this dependence and relation (9) it is found that  $U$  scales with  $B^{-1/2}$ . As already presented in Fig. 6,  $U$  scales empirically with  $B^{-1/2}$ , which is also compatible with Matsushita's analysis<sup>13)</sup>.

Consequently, we can conclude that flux creep is strongly affected by flux pinning and therefore microstructure. It is also clear that flux creep rate can be reduced by the introduction of effective pinning centers.

## 5. CONCLUSIONS

Through the comparison of critical currents and flux creep between the Y-Ba-Cu-O crystals with and without 211 inclusions, it can be concluded that the fine dispersion of 211 inclusions can help increase  $J_c$  in the Y-Ba-Cu-O system. We believe that non-superconducting phase can work as pinning centers even in high  $T_c$  oxide superconductors, although other defects can also contribute to flux pinning. It is also concluded that large flux creep rate is not inherent to Y-Ba-Cu-O superconductors even at 77K since the flux creep rate can be decreased by increasing flux pinning force through the microstructural control.

Acknowledgment We would like to thank Mr. K. Yamaguchi of Superconductivity Research Laboratory and Mr. M. Saga of Nippon Steel Corporation for transmission electron microscopic observation. We are also grateful to Mr. T. Oyama and Dr. Y. Shiohara of Superconductivity Research Laboratory for their help in sample preparation.

## REFERENCES

- 1) H. Kupfer, I. Apfelstedt, R. Flukiger, C. Keller, R. Meiner-Hirmer, B. Runtzsch, A. Turowski, U. Wiech and T. Wolf: "Intragrain junctions in  $YBa_2Cu_3O_{7-x}$  ceramics and single crystals" *Cryogenics* **29** (1989) 268.
- 2) R. B. Van Dover, E. M. Gyorgy, L. F. Schneemeyer, J. W. Mitchell, K. V. Rao, R. Puzniak and J. V. Waszczak: *Nature* **342** p. 55 (1989).
- 3) M. Murakami, M. Morita, K. Doi and K. Miyamoto: "A New Process with the Promise of High  $J_c$  in Oxide Superconductors", *Jpn. J. Appl. Phys.* **28** pp. 1189-1194 (1989).
- 4) H. Fujimoto, M. Murakami, S. Gotoh, T. Oyama, N. Koshizuka, Y. Shiohara, and S. Tanaka: "Melt Processing of YBaCuO Superconductors", *Proc. ISS 89* (Springer-Verlag, 1990) to be published.
- 5) R. I Coote, J. E. Evetts and A. M. Campbell: "Flux Line Pinning by Large Normal Particles in Type-II Superconductors", *Canad. J. Phys.* **50** pp. 421-427 (1972).
- 6) C. P. Bean: "Magnetization of Hard Superconductors", *Phys. Rev. Lett.* **8** pp. 250-253 (1962).
- 7) M. Murakami, M. Morita and N. Koyama: "Magnetization of a  $YBa_2Cu_3O_7$  Crystal Prepared by the Quench and Melt Growth Process", *Jpn. J. Appl. Phys.* **28** pp. L1125-L1127 (1989).
- 8) A. M. Campbell and J. E. Evetts: "Flux Vortices and Transport Currents in Type II Superconductors", *Adv. Phys.* **21** pp. 199-427 (1972).
- 9) M. Murakami, M. Morita and N. Koyama: "Flux Creep in High  $J_c$   $YBa_2Cu_3O_7$  Crystals", *Jpn. J. Appl. Phys.* **28** pp. L1754-1756 (1989).
- 10) M. Murakami, S. Gotoh, N. Koshizuka, S. Tanaka, T. Matsushita, S. Kambe and K. Kitazawa: "Critical Currents and Flux Creep in Melt Processed High  $T_c$  Oxide Superconductors", *Cryogenics* (1990) to be published.
- 11) T. Matsushita: *Jpn. J. Appl. Phys.* **20** p. 1955 (1981).
- 12) P. W. Anderson: "Theory of Flux Creep in Hard Superconductors", *Phys. Rev. Lett.* **9** pp. 309-311 (1962).
- 13) T. Matsushita: "Flux Pinning Mechanism and Critical Current Density in High-Temperature Superconductors", *Proc. ISS 89* (Springer-Verlag, 1990) to be published.